



Nanomaterials for CO₂ Capture from Gas Streams

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Abstract: Since CO_2 is an important component of gas emissions, its removal from gas streams is of the utmost importance to fulfill various environmental requirements. The technologies used to accomplish this removal are based mainly on absorption, as well as adsorption and membrane processing. Among the materials used in the above separation processes, materials in nano forms offer a potential alternative to other commonly used macromaterials. The present work reviews the most recent publications (2023) about CO_2 capture using different nanomaterials, and whilst most of these publications were dedicated to investigating the above, several presented data on the separation of CO_2 from other gases, namely nitrogen and methane. Furthermore, a number of publications investigated the recyclability of nanomaterials under continuous use, and just three of the references were about computational modeling; all others were experimental papers, and only one reference used a real industrial gas.

Keywords: CO₂; absorption; adsorption; membranes; nanomaterials; environment

1. Introduction

One of the main global concerns is related to the increasing concentration of CO_2 in the atmosphere, a consequence of the use of fossil fuels, boosting the greenhouse impact. There is no doubt that greenhouse gas emissions produce climate problems, resulting in challenges in reaching sustainable development. Greenhouse gas emissions are mainly a result of energy production (nearly 70% of global emissions). Many countries have considered the importance of reducing CO_2 emissions to fulfill environmental policies related to zero-carbon discharge, helping to reduce the presence of carbon in a number of sectors, with special interest in the energy sector. However, the time to achieve the transition from today's polluting energy production to a future of zero-emissions technology still seems long, and this is because there is a constant flux of information and developments to improve the capture of contaminant gases (CO_2 , H_2S , CH_4 , etc.) from gas streams in order to clean them and contribute to a better environment.

Among the technologies employed for this capture, and in the case of CO_2 specifically, absorption, adsorption, and membrane technologies are the most widely proposed. Several recent reviews on the utilization of these technologies have been published, including the use of membranes [1]; MXene-based membranes [2]; composite membranes [3,4]; microporous membranes composed of nanopores [5]; nanomaterials [6–11], and more specifically, graphene and its 2D nanomaterial derivatives [12]; nanomaterials derived with support from artificial intelligence (AI) [13]; kaolinite-based nanomaterials [14]; MXene nanoderivatives [15]; azobenzene-based supramolecular materials [16]; carbon-bearing nanomaterials [17]; nanobiotechnology using microalgae [18]; nanomaterials for catalyst-assisted solvent regeneration in absorption processes using amine [19]; and finally, some general reviews on technologies for CO_2 removal from gas streams [20–23].

This work reviews the most recent findings (2023) on the use of separation technologies to capture CO_2 from gas streams. The different studies are compiled based on the technologies used for capture.



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2. Nanomaterials and CO₂ Absorption

With quick kinetics, absorption techniques produce remarkable CO_2 removal (90%) from gases. The most-used absorbents to achieve this removal include amines, ammonia–water solutions, and alkali compounds. Both pre- and post-combustion processes can integrate these technologies into their general procedure. Besides physical absorption, chemical-driven absorption processes have three components: absorber, solvent, and stripper. Different pieces of equipment were used to capture CO_2 , i.e., spray columns, packed beds, rotating packed beds, bubble columns, and tray tower absorber layouts.

The absorption of CO_2 on Al_2O_3 /MeOH [24] was investigated, and the addition of a porous nickel metal foam increased CO_2 capture compared to pure MeOH; this increase was attributable to the forced bubble-breaking mechanism and the hydrodynamics in relation to the process. Moreover, there was a direct relation with the CO_2 absorption and the pressure and temperature used in the CO_2 capture process (Figure 1). For a constant pressure, increasing the temperature decreased the gas capture, whereas for a constant temperature, increasing the pressure produced a greater removal of CO_2 from the gas stream. Also, an increase (0.01–0.1 wt%) in the Al_2O_3 content is accompanied by an increase in the capture of CO_2 with the alcohol.



Figure 1. Approximate CO₂ removal in MeOH at various temperatures and pressures. Data from [24].

CO₂ capture by means of the addition of tetra-n-butyl ammonium bromide (TBAB) semiclathrate to graphite, forming a TBAB+graphite nanofluid, was investigated [25]. The TBAB+graphite nanofluid system increased CO₂ removal with respect to the use of the TBAB or TBAB+sodium dodecyl sulfate (SDS) solutions, with 0.2 wt% graphite nanoparticles being the best concentration for hydrate growth in the TBAB+graphite nanofluid.

A metal oxide solid acid catalyst for the catalytic regeneration of a CO_2 -rich 5 M monoethanolamine (MEA) solution at 90 °C was used [26]. After the adsorption process and the formation of MEACOO⁻, carbamate decomposition occurred:

$$MEACOO^{-} + H_3O^{+} \leftrightarrow Zwitterion \leftrightarrow MEA + CO_2$$
(1)

This was followed by protonated amine deprotonation and the formation of MEA, H_3O^+ , and H_2CO_3 . The utilization of $SO_4^{2-}/ZrTiO_x$ allowed for a 99% increase in the CO_2 desorption rate and a 43% enhancement after continuous cycles.

The performance of single-walled, carbon-deficient silicon carbide nanotubes $(Si_{12}C_{2-X}; X = 1; 2)$ in CO₂ capture was investigated [27]. The investigation concluded that $Si_{12}C_{11}$:Vc1 and $Si_{12}C_{10}$:Vc2 performed well for CO₂ removal and storage, surpassing the CO₂ capture efficiency of pristine SWSiCNT. The absorbent properties were due to the C vacancy effect of photoabsorption.

5,6-Dimethylbenzimidazole replaced 2-methylimidazole on the surface of ZIF-8 crystals via the shell–ligand exchange reaction (SLER) [28], improving the thermal stability of ZIF-8. At 40 °C, CO₂ uptake using ZIF-8-SLER-PLs increased 30% with respect to that of ZIF-8-PLs. CO₂ was loaded onto the modified absorbent via a physical absorption process.

It was demonstrated [29] that the use of nano-SiO₂, with varying particle size, in glycerol solution had little effect on CO₂ removal; this capture increased with the increase in solid mass in the 0.05–0.15 wt% range, which was attributed to a better gas–liquid mass transfer area. The use of SiO₂ increased CO₂ desorption as consequence of the presence of more nucleation sites and the heating rate of the base fluid, generating bubbles.

MFCs (MgFe₂O₄@ZIF-62) containing various magnetic nanoparticle dosages (1–6 wt% of magnetic nanoparticles to ZIF-62 mass) were used in conjunction with a compatible non-penetrating solvent to form a magnetic porous liquid [30]. The use of this absorbent material allowed continuous CO₂ capture and release for up to three cycles.

Due to certain operational and economic considerations, there is a relatively urgent necessity to find alternatives to the use of amines for CO₂ capture [31]; amino acid salts can be one green alternative to the use of these amines. Thus, potassium L-cysteine for CO₂ capture from natural gas was investigated. Its physicochemical properties were measured at different temperatures (25–60 °C) and salt concentrations (5–30 wt%). Experimental results, at 40 °C and 20 bar, showed an important increment of CO₂ loading, from 7 to 15 mmol CO₂/g amino acid, with the increase of 10–30 wt% in the solvent concentration. CO₂ loading was attributable to the following reaction:

$$CO_2 + alkanoamine \rightarrow carbamate$$
 (2)

while carbonate formation occurred via the following reaction:

carbamate +
$$H_2O \rightarrow amine + CO_3^{2-}$$
 (3)

By the use of 9.01 wt% tetra butyl ammonium bromide (TBAB) mixed with watersoluble hydroxylated multiwalled carbon nanotube (MWCNTol) material, the formation of CO_2 hydrate was investigated [32]. It was concluded that MWCNTols had negligible influence on the CO_2 hydrate generation. The use of nanoparticles such as graphene nanoribbons and MWCNTols reduced the induction time, whereas addition of various nanoparticle dosages to the TBAB solution increased the final gas consumption, with a maximum increase of 10.44% in the 9.01 wt% TBAB + 0.08 wt% GN system.

Absorption and conversion of CO_2 by an amino-acid-based nanotechnology was described [33]. CO_2 was captured as bicarbonate nanomaterials, whereas the amino acid structure governed the formation of bicarbonate nanomaterials. Amino acids presented higher CO_2 absorption capacity and faster kinetics compared to the use of 30 wt% monoethanolamine.

It was shown [34] that CO₂ absorption can be favored by the use of low-transitiontemperature mixtures. This technology uses nanofluids and the addition of, i.e., TiO_2 nanoparticles. The CO₂ absorption efficiency was enhanced by a factor of 1.35 when the nanoparticle dose was 0.6 kg/m³ and presented a particle size of 10 nm. As shown in Table 1, there was an optimal concentration of TiO_2 nanoparticles in the CO₂ absorption process. At a fixed size of the nanoparticles, the CO₂ absorption efficiency increased with the decrease in the diameter of nanoparticles.

Gold nanoparticles were decorated with 1,5,7-triazabicyclo [4.4.0] dec-5-ene and dispersed into methanol in order to capture CO_2 [35]. The photocatalyst consisted of two parts: (i) an organic shell responsible for CO_2 capture, and (ii) a plasmon-active metal nanoparticle core for activation of captured CO_2 and its involvement in the cycloaddition reaction. Results showed the efficiency of the procedure even at the temperature of -40 °C.

The mixture formed by NaP zeolite nanocrystals and 1-dodecyl-3-methylimidazolium chloride ($[C_{12mim}][Cl]$) ionic liquid was used for CO₂ removal in an isothermal high-pressure cell equipped with magnetic stirring [36]. Under various experimental conditions,

Methyl-diethanolamine-based Fe₃O₄ improved CO₂ absorption compared to methyldiethanolamine-based CuO, ZnO, and SiO₂, whereas CuO nanoparticles presented higher efficiency for CO₂ removal from gas-loaded absorbent [37].

Using 2-methylimidazole zinc salt (ZIF-8) modified by tetraethylenepentamine (TEPA), which provided pores, and 1-ethyl-3-methylimidazolium bis(trifluoro-methanesulfonyl)-imide ([EMIm][NTf₂]) ionic liquid, used as a sterically hindered diluent, an amine-functionalized type III porous liquid was formed [38]. It was found that with 30TEPA@ZIF-8 nanoparticles, the best CO₂ absorption capacity was obtained. Moreover, the CO₂ absorption loading of 0.124 mmol/g presented by 5-30TEPA@ZIF-8/[EMIm][NTf₂] was 4.43 times higher than the value obtained by the use of 5-0TEPA@ZIF-8/[EMIm][NTf₂], whereas a 745% increase of the absorption rate was reached.

A mixture of 26 mol% CO_2 and 74 mol% CH_4 was used to investigate the separation of both gases [39]. Silica nanoparticles, in KOH medium, modifying the surface of (3aminopropyl) teriethoxysilane (APTES) were utilized as additives. The best results were derived with silica and KOH-bearing nanofluids; with these components, improvements of 36% (gas consumption), 29% (separation factor), and 38% (recovery factor) resulted with respect to the use of pure water.

 CO_2 geological sequestration by the use of silica aerogel nanofluid was investigated [40]. Using this material, non-dissolved CO_2 molecules were captured in the nanopores of the silica aerogel nanoparticles, increasing the solubility of CO_2 in the aqueous phase. Aerogel nanoparticles adsorbed at the CO_2 -brine interface reduced the interfacial tension.

Table 2 summarizes the nanomaterials and objectives of the above references.

Cp, kg/m ³	10 nm	25 nm	60 nm
0	1	1	1
0.2	1.21	1.16	1.15
0.4	1.23	1.22	1.21
0.6	1.35	1.30	1.17
0.8	1.20	1.24	1.12
1.0	1.15	1.10	1.10

Table 1. Approximate enhancement factor (E) values at various nanoparticle loadings and diameters.

Temperature: 30 °C. Data from [34].

Table 2. Summary of the use of nanomaterials in the absorption of CO_2 .

Ref.	Nanomaterial	Objective
[24]	Porous Ni foam	CO_2 capture
[25]	TBAB+graphite nanofluid	CO ₂ removal
[26]	$SO_4^{2-}/ZrTiOx$	CO ₂ desorption rate
[27]	Silicon carbide nanotubes	CO_2 capture/storage
[28]	ZIF-8+DMBI	Improve ZIF-8 thermal stability
[29]	Nano-SiO ₂ and glycerol	CO_2 desorption
[30]	Nanomagnetic porous liquid	Continuous use up to 3 cycles
[31]	K-L-cysteine	Substitution of amines
[32]	TBAB+MWCNTs	Formation of CO ₂ hydrate
[33]	Amino-acid-based technology	CO ₂ conversion to bicarbonate
[34]	Nanofluids+TiO ₂ nanoparticles	CO ₂ removal
[35]	Gold nanoparticles	CO ₂ capture
[36]	Zeolite nanocrystals+ionic liquid	CO ₂ removal
[37]	MDA-Fe ₃ O ₄	CO_2 removal
[38]	Modified ZIF-8 salt	CO ₂ capture
[39]	APTES+silica nanoparticles	CO_2/CH_4 separation
[40]	Silica aerogel nanofluid	CO_2 geological sequestration

TBAB: tetra-n-butyl ammonium bromide. DMBI: 5,6-dimethylbenzimidazol. MWCNTs: multiwalled carbon nanotubes. MDA: methyl diethanolamine. APTES: (3-aminopropyl) teriethoxysilane.

Though it was difficult to compare results due to the different experimental conditions used in each reference and how they influenced CO_2 uptake, strikingly different values such as 7–15 mmol CO_2/g and 0.25 mmol CO_2/g from references [31,38], respectively, were found.

3. Nanomaterials and CO₂ Adsorption

Adsorption processing involves the use of a solid material on which CO_2 (and other gases and solutes) is captured by means of physical or chemical processes or a combination of both. Key parameters to yield the best adsorptive properties of the materials are: porosity, pore size, operational stability, presence of reactive groups towards CO_2 adsorption, etc., whereas the equipment used is usually described as a packed or fluidized bed.

The fabrication of carbon nanofibers (CNFs) via biaxial electrospinning was investigated [41]. Polymethylmethacrylate (PMMA) and polyacrylonitrile (PAN) were used as core and shell precursors, respectively. Further, Co_3O_4 nanoparticles were included in the PAN shell, increasing its roughness and surface area. The uniform distribution of Co_3O_4 resulted in a better flexibility of the hollow carbon nanofiber material (HCNF-Co), providing more vacant oxygen sites to increase CO_2 adsorption loading. HCNF-Co nanofibers exhibited CO_2 capture uptake of 3.28 mmol/g at 25 °C. Experimental results indicated that HCNF-Co had remarkable CO_2 selectivity (S = 26) over N₂.

Heterojunctions of Co_3O_4 with different morphologies and modified carbon nitride (CN) were investigated in order to optimize their properties to degrade CO_2 under UV-visible irradiation [42]. A solvothermal synthesis was used to fabricate the cobalt oxide from metal–organic framework structures, yielding ultrathin 2D Co_3O_4 nanosheets (Co_3O_4 -NS). These nanosheets presented improved photocatalytic properties compared to those of the bulk Co_3O_4 /CN composites. CO_2 reduction was improved due to (i) the match of the planar surface of CN and the 2D structure of Co_3O_4 -NS, which resulted in a larger interface, and (ii) improvement in charge carrier lifetime.

The authors of [43] described the utilization of 2D nanomaterial MXenes and activated carbon (AC) to form sandwich-type materials and nanocomposites for CO₂ adsorption using a fixed-bed column. These investigations included CO₂ breakthrough measurements at a fixed 15% CO₂ concentration, with an inlet flow rate at 200 mL/min and temperatures in the 25–55 °C range. The highest CO₂ adsorption load (near 9 mg/g) was yielded with AC/MXene sandwich adsorbent at 25 °C, which was nearly a 37% improvement in CO₂ adsorption capacity over the use of pristine AC. AC/MXene sandwich-type nanomaterials can be used, with a small loss of their CO₂ adsorption uptake, under various cyclic experiments.

ZIF-8 hollow nanospheres, for selective CO₂ separation and storage, were developed [44]. The optimum hollow ZIF-8 nanosphere material, with a uniform size distribution (Figure 2), had a CO₂ adsorption uptake of 2.24 mmol/g at 0 °C and 1.75 bar, selective (12.15) CO₂/N₂ separation, 1.5–1.75 wt% CO₂ storage capacity, and a reasonable stability, up to four CO₂ adsorption cycles, at 25 °C.

A heterogeneous catalyst comprising silver nanoparticles and a porous N-heterocyclic carbene polymer (Ag@POP-NL-3) was developed [45]. This nanomaterial has a regular distribution of silver nanoparticles and nitrogen activation groups. The catalyst presented good properties for the selective adsorption and activation of CO_2 , allowing the conversion, under mild conditions, of low CO_2 (30 vol%) concentrations, as presented in lime kiln waste gas, into cyclic carbonate. CO_2 was loaded onto the adsorbent by carboxylative cyclization of the gas with propargylic alcohols also present in the system.

The adsorption uptake of CO₂ on NaY@polyacrylate matrix was increased by 17.9% while H_2O adsorption uptake decreased by 36.6% compared to pristine NaY [46]. In addition, H_2O adsorption was reduced by 54.8% after adding ZIF into composites.

The authors of [47] described a maximum CO_2 loading (0.75 mmol/g) on triethylaminedoped rice husk silica nanoparticles, with an average increase in CO_2 adsorption with the increase (1 to 5 wt%) in the amine loading on the surface modifiers. Amine loadings greater than 5 wt% produced agglomeration of the particles which is detrimental with respect to CO_2 capture. CO_2 uptake corresponded to the Langmuir isotherm model.



Figure 2. Influence of synthesis time on the average diameter of soft template hollow ZIF-8 nanospheres. Surfactant/oil ratio: 75 g/L. Data from [44].

The use of Zn-N pillar MOFs resulted in: CO_2 capture of 3.82 mmol/g (25 °C and 101 kPa), a selectivity CO_2/N_2 factor of 132, and stable structure (no change after exposure to 1000% RH environment for seven days) [48].

The performance of graphene oxide (GO)-coated zinc tetraphenylporphyrin (ZnTPP/GO) nanocomposites in the photocatalytic degradation of CO_2 was investigated [49]. The encapsulation of GO in ZnTPP nanocrystals promotes CO_2 adsorption, interfacial reaction, and stability and accelerates the separation of photoinduced carriers on ZnTPP (0.1 ps vs. 425.9 ps), the transportation from ZnTPP to GO (2.3 ps vs. 83.6 ps), and their final enrichment on GO.

A porous ZIF-11@ZIF-8 core–shell composite structure metal–organic framework was fabricated using the solvent-assisted linker exchange (SALE) procedure [50]. Adsorptions at 25 °C and equilibrium pressures up to 4 bar showed an increase (near 100%) in CO₂ adsorption uptake of ZIF-11@ZIF-8 nanoparticles (8.21 mmol/g) compared to the pristine ZIF-11 (4.35 mmol/g). Experimental results on gas uptake fitted well with the Langmuir isotherm equation. CO_2/N_2 and CO_2/CH_4 selectivities also increased by 131% and 92%, respectively.

Activated carbon (AC) was synthesized from date fruit seeds and chemically activated with KOH to improve CO_2 loading [51]. From thermogravimetric analyses, 94% and 67% higher average CO_2 capture loads were measured for KOH-promoted ACs compared to the original adsorbents. The activated carbon improved its fluidization by the use of hydrophobic silica nanoparticles (NPs). The SiO₂-decorated (2.5 wt%) modified ACs had a 45% higher bed expansion ratio, which was associated with the absence of bubbles and a homogeneous fluidized regime.

Mesoporous CeO₂, ZrO₂, and Ce-Zr composite nanoparticles with a large surface area were fabricated using the hydrothermal template-assisted synthesis procedure, and CO₂ adsorption properties of these materials were investigated under equilibrium and dynamic operations [52]. Better CO₂ adsorption was yielded for Ce-Zr nanomaterial due to the presence of strong O^{2−} base sites and many surface oxygen species. After five adsorption/desorption cycles, the composites presented a reasonable stability with a slight decrease in CO₂ adsorption uptakes in dry flow and in the presence of water vapor. Treatment via surface N_2 plasma of zinc porphyrin (ZnTCPP) ultrathin nanosheets induced nitrogen vacancies (NVs) and resulted in a material with photocatalytic CO₂ reduction activity and selectivity [53]. It was shown that the photocatalytic activity of NVs-ZnTCPP can be attributed to nitrogen-vacancy-induced spin polarization by reducing the reaction barriers and inhibiting the recombination of photoexcited carriers.

It was reported [54] that CO_2 uptake (11.8 mmol/g (78% total adsorption)) after four cycles on a MOF-derived nano-CaO (average size of 100 nm) was due to high stability produced in the final material by the change in the original fiber-bundle-like MOF structure to nanosheets, and further to regular CaO spheres. CO_2 uptake onto the adsorbent corresponded to the following equation:

$$CaO + CO_2 \leftrightarrow CaCO_3 \Delta H^0_{298} = \pm 178 \text{ kJ/mol}$$
(4)

Lewis base and dual hydrogen bond donor (HBD) units were integrated into an organosilicon precursor, and triazine and hydrazo site co-modified periodic mesoporous organosilicas (THPMOs) were prepared via a hydrothermal self-assembly method [55]. The THPMOs had BET surface areas in the 699–876 m²/g range and low-pressure CO₂ adsorption loadings at 0 °C. If combined with tetrabutylammonium iodide (TBAI) ionic liquid, the mixture promoted the model cycloaddition of CO₂ in an effective form, with the gas fixed to epoxides.

In [56], nano-TiO₂ was added to cement pastes to investigate its performance regarding the CO₂ uptake rate. Prismatic samples with dimensions of $16 \times 4 \times 4$ cm of 0.5 water/binder cement paste with and without nano-TiO₂ particles were used. CO₂ uptakes showed that nano-TiO₂ addition improves the CO₂ uptake rate of cement pastes, changing the pore structure and allowing the removal of more CO₂ at lower gas concentrations. CO₂ loaded similarly to Equation (4), but Ca(OH)₂ reacted with the gas to form CaCO₃ and water.

MIL-101(Cr)-NH₂ has higher CO₂ adsorption capacity than MIL-101(Fe)-NH₂ [57], whereas the adsorption of methane and nitrogen by MIL-101(Cr)-NH₂ is lower than the adsorption of these gases by MIL-101(Fe)-NH₂, leading to a higher selectivity of CO₂ over the two gases for MIL-101(Cr)-NH₂. At elevated temperature and pressure, the chemisorption mechanism is predominant, which is attributable to the performance of amines, which adsorbed more CO₂ at these higher temperatures and pressure. Gas adsorption was explained by the use of a hybrid equation between Langmuir and Khan models:

$$q = q_s \frac{P}{(1+P)^n} + Q \frac{P}{(1+P)^n}$$
(5)

where q_s , Q, and n represent the parameters of the model, with P being the pressure of the vapor phase at the equilibrium.

Three ceria nanoparticles, of C-CeO₂ (cubic), O-CeO₂ (orthorhombic), and S-CeO₂ (spherical) morphologies, were investigated for CO₂ adsorption [58]. Further, the best nanoparticle morphology (C-CeO₂) was doped with 1 wt% of Ni and Pd (C-CeNiPd) to investigate its performance in CO₂ adsorption. CO₂ adsorption increased in the order C-CeO₂ > O-CeO₂ > S-CeO₂ in the 30–200 °C temperature range (Table 3). C-CeO₂ material doped with 1 wt% Ni and Pd (C-CeNiPd) presented the best results (34% CO₂ adsorption) in relation with pristine C-CeO₂ (near 24%). CO₂ adsorption decreased, in all the systems, under steam injection.

A furan-derived di-functional epoxy monomer (5-hydroxymethyl-2-furaldehyde-3,5diamino-1,2,4-triazole) and a curing agent (5,5'methylenedifurfurylamine) were used to fabricate multifunctional carbon foams, which presented a CO₂ capture uptake of 4.72 mmol/g at 0 °C. This capacity was attributable to the large surface area (472 m²/g) presented by the adsorbent [59].

т, °С	C-CeO ₂	O-CeO ₂	S-CeO ₂
30	7	6.5	5
50	6.8	6.2	5.8
100	6.5	5.6	4.6
200	4.4	3.8	2.7

Table 3. Approximate CO_2 adsorption capacities (mmol/g) at various temperatures.

Pressure: 3 MPa. Data from [58].

Mesoporous cobalt oxide (Co₃O₄) nanoparticles (NPs) were fabricated in a medium containing choline chloride/ethylene glycol by the ultrasound-assisted method [60]. The results show that the adsorptive properties of Co₃O₄ NPs synthesized in the choline chloride/ethylene glycol medium improved when compared with Co₃O₄ NPs synthesized in water. CO₂ uptake corresponded well to the Langmuir isotherm, with the gas loading onto the adsorbent due to interaction, via the carbon atom, of CO₂ molecules with the oxygen atom of the Co=O groups.

A ZIF-8@Zn-MOF-74 metal–organic framework presenting, among other features, a core–shell structure was formed [61]. The CO₂ adsorption measurements were conducted at 35 °C and 4 bar. The CO₂ uptake (3.27 mmol/g) of ZIF-8@Zn-MOF-74 nanoparticles improved by about 64% in relation to the capacity shown by pristine ZIF-8 (2 mmol/g). CO₂ adsorption corresponded to the dual-mode adsorption equation, which included the Langmuir isotherm and Henry's law:

$$C(P_e) = K_d + \frac{C_s b P_e}{1 + b P_e}$$
(6)

in the above equation, $C(P_e)$ represents the amount of equilibrium gas uptake, K_d the Henry's law constant, C_s the Langmuir capacity constant, b the gas–site affinity constant, and P_e the gas equilibrium pressure. The use of these nanoparticles produced an increase (85% for the pair CO_2/N_2 and 74% for the pair CO_2/CH_4) in the selectivity values for these gases.

CeO₂ and CeO₂-ZrO₂ (75% CeO₂) mixed oxide were investigated to adsorb CO₂ under static and dynamic operations [62]. Commercial ceria materials, having similar textural and structural characteristics, promoted, after CO₂ uptake, the formation of carbonate-like surface species, which resulted in nearly the same CO₂ capture efficiency when dynamic and static operations were conducted. The synthesized CeO₂ material having a 30% larger surface area presented a long mass transfer zone in the breakthrough curves. On dynamic operation and with the same surface area as the synthesized CeO₂, CeO₂-ZrO₂ oxide had a greater CO₂ removal capacity (136 μ mol/g) resulting from the increase in the CO₂ adsorption sites (including defects) presented in this material.

Various metalloporphyrin- and triazine-integrated nitrogen-rich frameworks ($M \subset PPTFs$ (M represents a metal)) were formed and characterized, and their behavior in the adsorption of CO₂ and their catalytic properties to convert CO₂ to cyclic carbonates, via CO₂ cycloaddition to epoxide, were investigated [63]. Results showed that $Zn \subset PPTF15$ presents two functions: CO₂ adsorption and its synergistic conversion. CO₂ uptake reached 2315 µmol/g and 1883 µmol/g at 0 and 25 °C, respectively. In conjunction with the use of tetrabutylammonium bromide (TBAB) ionic liquid, and 60 °C and 0.1 MPa CO₂ pressure, $Zn \subset PPTF15$ reached 96% yield of chloropropene carbonate with 99% selectivity after a reaction time of 15 h. Furthermore, $Zn \subset PPTF15$ maintained its adsorptive properties after up to five cycles.

A series of zinc- and copper-based MOFs and carbon-supported Cu catalysts were constructed and used to the cycloaddition, carboxylation, and carboxylative cyclization reactions with CO₂; in the process and as targeting products, cyclic carbonates, carboxyl acids, and oxazolidinones were formed [64].

Cellulose-templated CaO-based pellets were used to remove CO_2 , and after twenty cycles the gas uptake was 7.3 mmol/g [65]. This material accounted for an increase of 133% in comparison with the use of the raw adsorbent without steam reactivation.

Polyethyleneimine-based nanoparticles were used as bicarbonate for CO_2 capture, increasing the CO_2 that reacts with the RuBisCO enzyme, which resulted in a twenty percent increase in the 3-PGA production in in vitro experiments [66]. Loading experiments concluded that, in vivo, the capture of CO_2 was maintained, thus widening its reloading with atmospheric CO_2 while *in planta*. In the loading process, amine groups reacted with water to form $-NH_3^+OH^-$ species, which further reacted with CO_2 to form $-NH_3^+OCOOH$ species. The same reactions occurred when secondary and tertiary amines were used instead of primary ones, as above.

By the use of a supported Ni–CaO composite catalyst for CO₂ capture, it was found that in the CO₂ conversion (96.5% at 650 °C), the balance between size and loading density of the nickel nanoparticles on calcium oxide controlled the adsorptive/catalytic interface [67].

A fiber-based CO₂ adsorbent, using in situ growth of ZIF-8 on 3D fibrillar lyocell/feather non-wovens (ZIF-8@LFNW), was fabricated [68]. The 12 wt% ZIF-8@LFNW material presented the best (4.46 cm³/g) CO₂ uptake in a high- and low-CO₂-concentration environment. CO₂ uptake was attributable to physical and chemical adsorption processes. This nanomaterial maintained its properties after various adsorption/desorption cycles.

Dual-pore carbon nitride (DP-CN) materials containing different ethylenediamine (EDA) to carbon tetrachloride (CTC) ratios, various potassium hydroxide dosages, and nanosilica were synthesized [69]. DP-CN-1-4 (where 1 represents the EDA to CTC mass ratio and 4 is the KOH to M-CN-1 mass ratio) was the material presenting the best adsorption results, with CO₂ uptakes of 8.3 mmol/g (1 bar), 16.9 mmol/g (10 bar), and 22.9 mmol/g (30 bar). This material separated CO_2/N_2 and CO_2/CH_4 (Table 4), being stable after numerous adsorption–desorption cycles (100% CO₂ adsorption in the first cycle versus 96.5% after the fifth cycle).

Gas Mixture	0.1 Bar	0.5 Bar	1 Bar
CO ₂ /N ₂ 50:50	32	51	60
CO ₂ /N ₂ 15:85	26	40	45
CO ₂ /CH ₄ 50:50	23	32	36
CO_2/CH_4 15:85	20	25	28

Table 4. Approximate selectivity values of DP-CN-1-4 under different pressures and gas mixtures.

Selectivities calculated using the ideal adsorbent solution theory (IAST). Data from [69].

In [70], a mimic carbonic anhydrase, using 2D nitrogen-doped graphene with Zn nanoclusters (Zncluster/NG), was fabricated. Tuning the pyrolysis conditions, the enzymemimicking activities of Zncluster/NG were regulated, with Zncluster/NG-900–800NH₃ nanozyme being the material having the best activity properties, with CO₂ hydration forming bicarbonate groups. This nanomaterial can be used in a wide range of temperatures and pH values, showing ten times greater catalytic activity at 75 °C than at room temperature.

A dual-functional catalyst–sorbent for CO_2 capture from air and in situ solar-driven conversion to produce clean fuels was developed [71]. At 25 °C and an initial CO_2 concentration of 500 ppm, the material presented 0.38 mmol/g of CO_2 loading, with a CO_2 conversion rate of 95%. In the conversion step, injection of methane transformed loaded CO_2 into CO and hydrogen.

AgInS₂ QDs-MoS₂/GO (AIS-MS/GO) composites using the interfacial coupling of AgInS₂ QDs (AIS), hierarchical MoS₂, and ultrathin GO were formed [72]. Under optimal conditions, the concentrations of CO and CH₄ in the CO₂ reduction products were 52.6 and 28.5 μ mol/g, respectively.

By the use of machine learning, the CO₂ adsorption capacity onto a carbon-based material in optimized conditions of pressure, temperature, micro- and mesopore volumes,

specific surface area, and carbon, oxygen, hydrogen, and nitrogen concentrations was predicted [73].

The regeneration of CO_2 adsorbents, based on an ion exchange mechanism, was proposed [74]. Using a concentrated OH⁻ solution, a quaternary ammonium (QA)-based ion exchange resin can be changed from a CO_3^{2-} to OH⁻ cycle, promoting CO_2 uptake (as carbonate or bicarbonate species) from ambient air. In this OH⁻ cycle, the loading capacity was 1.85 mmol/g, surpassing 0.88 mg/g from the CO_3^{2-} cycle. Experimental results indicated that the alkaline solution/HCO₃⁻ inlet ratio, concentrations, and external diffusion were the key parameters to influence the desorption rate of CO_3^{2-} attributed to the ion exchange mechanism.

A solvothermal procedure to fabricate a series of urea-functionalized ionic organic polymer (UIP) materials (UIP-1, UIP-2, and UIP-3) was used [75]. UIP-2, formed from the reaction of 1,3,5-tribromomethyl benzene, 4-amino pyridine, and 4,4'-diphenylmethane diisocyanate, presented remarkable catalytic efficiency in the CO₂ cycloaddition process, via fixation to epoxide, with a 92.65 cyclic carbonate yield using the following experimental conditions: epichlorohydrin (ECH) (10 mmol), UIP-2 (0.10 mmol), reaction at 70 °C for 16 h with 1 bar of CO₂.

Oriented attachment, as a procedure for the generation of ordered 1D/2D NiZn layered double hydroxide (Ni-Zn LDH) heterostructures, was developed [76]. The oxygen-deficient 1D NiZn-LDH material, useable for CO₂ adsorption, was dispersed on the 2D platform. Under visible light exposure, the oriented 1D/2D NiZn-LDH heterostructure degraded CO₂, reaching a 16.95 μ mol/g·h CO rate and complete CO selectivity.

The topological transformation of 2D layered double hydroxides allowed the fabrication of 2D transition metal oxide (TMO) nanomesh of ultrathin thickness [77]. A 2D Co/NiO-2 nanomesh presented better photocatalytic CO₂–syngas conversion efficiency, i.e., VCO of 32,460 μ mol/g·h CO, with VCO about 7.08 and 2.53 times the values presented by NiO and 2D Co/NiO-1 nanomesh, respectively.

Several ZnO@ZIF composites were prepared to investigate CO_2 capture and photocatalytic reduction [78]. The adsorption of CO_2 depended on S_{BET} , pore volume, and size, increasing at higher S_{BET} , pore volume, and with a decrease in the pore size (Figure 3). CO_2 was converted to CO due to the synergism between ZnO NFs and ZCZIF, that increased light trapping, including visible (400–700 nm) and UV (200–400 nm) absorption bands.



Figure 3. Dependence of CO₂ adsorption with the adsorbent pore size. Data from [78].

Table 5 summarizes the nanomaterials and objectives of the above references.

Ref.	Nanomaterial	Objective
[41]	Carbon nanofibers+Co ₃ O ₄	CO_2 uptake and CO_2/N_2 separation
[42]	Carbon nitride+ Co_3O_4	CO_2 photodegradation
[43]	2DMXenes+activated carbon	CO_2 uptake in fixed-bed column
[44]	ZIF-8 hollow nanospheres	CO_2/N_2 separation and CO_2 storage
[45]	Ag nanoparticles+carbene polymers	CO_2 uptake and conversion to CO_3^{2-}
[46]	NaY@polyacrylate matrix	CO_2/H_2O separation
[47]	Doped rice husk silica nanoparticles	CO ₂ uptake
[48]	Zn-N pillar MOFs	CO_2 uptake and CO_2/N_2 separation
[49]	GO+Zn/TPP nanocomposite	CO_2 capture
[50]	ZIF11@ZIF-8 structures	CO_2/N_2 , CO_2/CH_4 separation
[51]	Activated carbon+nanosilica	CO ₂ removal
[52]	Ce-Zr nanocomposites	CO ₂ loading
[53]	Zn/TCPP nanosheets	Intensify photocatalytic CO ₂ reduction
[54]	MOF-derived nano-CaO	CO_2 uptake up to 4 cycles
[55]	THPMOs	CO ₂ adsorption at 0 °C
[56]	Nano-TiO ₂ +cement pastes	CO_2 capture
[57]	MIL-101(Cr)-NH ₂	CO_2/N_2 , CO_2/CH_4 separations
[58]	Ceria nanoparticles	CO ₂ uptake
[59]	Carbon foams	CO_2 capture at 0 °C
[60]	Co_3O_4 nanoparticles	CO_2 capture
[61]	Metal frameworks	CO_2/N_2 , CO_2/CH_4 separations
[62]	Ceria derivatives	Static and dynamic CO ₂ capture
[63]	Integrated N ₂ -rich frameworks	CO ₂ uptake and catalytic conversion
[64]	Zn, Cu-based MOFs	CO_2 adsorption and catalytic conversion
[65]	Cellulose-CaO-based pellets	CO ₂ adsorption after twenty cycles
[66]	PEI nanoparticles	CO_2 capture
[67]	Ni-CaO composite	CO_2 capture and catalytic conversion
[68]	ZIF-8 derivative	CO_2 capture
[69]	Dual-pore carbon nitride	CO_2/N_2 and CO_2/CH_4 selectivity
[70]	Nanozymes	CO_2 capture and catalytic conversion
[71]	Dual catalyst-adsorbent	CO_2 uptake and conversion
[72]	Ag-In-Mo composites	Reducing CO and CH_4 in CO_2 products
[73]	Carbon-based material (MLP)	Prediction of CO ₂ uptake
[74]	Various chemicals	Regeneration of CO_2 adsorbents
[75]	Urea derivatives	Catalytic efficiency in CO_2 cycloaddition
[76]	N1-Zn heterostructures	CO_2 capture, photocatalytic conversion
[77]	2D metal oxide nanomesh	Photocatalytic CO_2 -syngas conversion
[78]	ZnO@ZIF nanocomposites	CO_2 capture and photocatalytic reduction

Table 5. Summary of the applications of nanomaterials in CO₂ adsorption.

MOF: metal–organic framework. TPP or TCPP: tetraphenylporphyrin. THPMOs: triazine and hydrazo site co-modified periodic mesoporous organosilicas. PEI: polyethyleneimine. MLP: machine learning prediction.

As one can see from the above table, some of the references investigated CO_2 uptake onto the nanomaterials together with photocatalytic gas degradation or gas catalytic conversion. Also, some of the investigations were dedicated to the separation or selectivity of CO_2 from other gases (N₂ and CH₄). Two of the references investigated CO₂ capture at 0 °C, and two others used dynamic experiments (columns). It is also worth noticing here that just one investigation was dedicated to the computational modeling of the adsorption process, and none of the references appearing in Table 4 used real gas samples but mimicked gases streams in their studies. Though it was difficult to compare CO_2 uptakes onto the various nanoadsorbents, basically due to the different experimental conditions used in the experimentation, Table 6 shows a series of (absolute) CO_2 loadings onto the respective nanoadsorbents.

From the data presented in the above table, the great disparity between the values representing the maximum CO_2 uptake onto the respective nanomaterials is clear.

Ref.	Limit	CO ₂ Uptake, mmol/g
[69]		22.9
[50]	Upper	8.2
[65]	* *	7.3
[47]		0.75
[71]	Low	0.38
[62]		0.13

Table 6. Selected maximum CO₂ uptakes onto various adsorbents.

CO₂ uptake may be dependent on diverse factors, i.e., temperature, pressure, initial CO₂ concentration.

4. CO₂ Capture by Membrane Technologies

The usefulness of membranes in separation methodology relies on the permeation rates of each gas component across the membrane, affording the selective separation of one gas from another. They offer a greater novelty compared with other established separation technologies, including absorption and adsorption procedures. Due to a number of their operational characteristics, membranes are considered a challenge for the separation of the various components of a gas stream. In the case of CO₂ separation, the membranes used can be broadly classified as inorganic, polymer, and composite membranes.

Membranes allow the selective passing of solutes across their structure. Membranebased separation has different uses in different industrial fields; particularly, the increase in gas separation applications to reach the goal of carbon neutrality and a circular economy makes membrane-based separation a suitable approach.

Hollow fiber membrane modules are often used to investigate gas permeation across membrane fibers. Under operational conditions, CO₂ (or any gas) flows on the shell side and diffuses across the membrane fibers to the tube side; here, the gas is normally absorbed by a receiving phase. These modules operate under different operational forms, with the counter-current form usually preferred over the co-current form, since it gives a better contact between gas and liquid phases. Figure 4 shows this counter-current configuration, and Figure 5 represents the mass transfer occurring in the module.



Figure 4. Commercial hollow fiber membrane module to investigate permeation phenomena. Phases enter the module in counter-current operational form. Module length: 28 cm. Fiber length: 15 cm. Effective membrane area: 1.4 m^2 .



Figure 5. Mass transfer in the hollow fiber module. Curved black lines represent the membrane pores. Vertical arrows: CO₂ flux. Horizontal arrows: Phase fluxes in counter-current mode.

A model based on the Bayesian regulation algorithm was used to simulate different CO_2 permeabilities in PMP/ZnO, PMP/Al₂O₃, PMP/TiO₂, and PMP/TiO₂-nanotubes (PMP = poly(4-methyl-1-pentene)) membranes [79]. The modelization results presented an absolute average relative deviation lower than 5.5%, mean absolute error of 6.87, and correlation coefficient higher than 0.99470. The mixed matrix of PMP/TiO₂-nanotubes is the best membrane for CO_2 separation.

A membrane that presented convenient performance for CO_2 separation and at the same time a reasonable cost was produced [80]. Thus, 4A-zeolite together with Pebax-1657 polymer was used to create the membrane and, by the change in the operational conditions of the process, the permeability and selectivity of the membrane were measured. The CO_2 selectivity compared to N₂, O₂, and CH₄ improved by 53, 67, and 75%, respectively.

The capture of CO_2 on various polymer-bearing nanoparticles such as poly(ether-blockamide) was investigated [81]. PEBA2533 dissolved in ethanol was used to make a PEBA membrane, which was doped with tetraethylenepentamine (TEPA) or hexadecylamine (HDA). The best CO_2 capture results were obtained when the membrane contained 2.5% TEPA or 1% TEPA and HAD.

The authors of [82] investigated the formation of high-aspect-ratio (30/1) KAUST-8 nanosheets with a CO₂ uptake of 60 cm³/g. This resulted in a loading of 30 wt% in the crosslinked poly(ethylene oxide) membrane, at the same time increasing CO₂ permeability (280%) and CO₂/N₂ selectivity (30%) in the mixed 50/50 vol gas permeation measurements.

By the use of UiO-66 nanofillers and polyetherimide (PEI), mixed matrix membranes were fabricated [83]. When compared with pristine PEI membranes, mixed membranes, containing metal–organic framework nanoparticles, have better H₂ permeation and H₂/CO₂ separation factor, i.e., permeation values of 14.6 and 5.5 Barrer were reached for H₂ and CO₂ at 10 wt% nanofiller concentration and 4.0 bar of applied pressure.

 CO_2 -philic zirconium-based MOF NPs (Zr BDC or UIO-66) were made, decorated with I-lysine amino acids, to be incorporated into a chitosan (CS) polymer matrix [84]. It was shown that the membranes containing 7 wt% of lysine-conjugated Zr BDC (lys-c-Zr BDC) NPs with a 4 µm active layer thickness had better adsorptive yields than pristine CS and the Zr BDC-embedded CS MMM (Table 7). The composite lys-c-Zr BDC-incorporating CS mixed membrane showed a CO₂ permeance of 34.9 GPU and CO₂/N₂ separation factor of 29.4 (dry conditions) and a CO₂ permeance of 135.2 GPU and a steady CO₂/N₂ separation factor of 71.5 (swollen conditions at 85 °C and 0.221 MPa feed pressure). MOF nanoparticles in the membranes provided the high porosity and surface area, which contributed to increasing CO₂ permeability, while the selectivity was due to the amine functional group of I-lysine.

Material	Material wt%	CO ₂ Permeation	N ₂ Permeation
	0	14	<0.5
7 000	3	25	< 0.5
ZrBOC	7	79	< 0.5
	10	65	<0.5
	0	14	<0.5
Lys-c-ZrBOC	3	40	< 0.5
	7	135	< 0.5
	10	132	<0.5

Table 7. Approximate CO₂ and N₂ permeation values (GPU) at various ZrBOC and lys-c-ZrBOC loadings.

Pressure: 0.221 MPa. Temperature: 85 °C. Sweep/feed mixture flow ratio: 1.67. Data from [84].

In order to assist CO₂ permeation across a graphene oxide membrane, melamine was introduced into the graphene oxide layer [85]. Using this new membrane, CO₂ permeance reached 68.02 Barrer, and at the same time CO_2/N_2 separation reached the value of 37.75, twice the value presented by graphene oxide membranes. The amine function from melamine, inserted into the graphene oxide membrane, contributed to CO₂ permeation and CO_2/N_2 selectivity.

In-situ-formed PEO networks with silica nodes were introduced into Pebax-1657, resulting in a semi-interpenetrating network structure [86]. This material increased CO₂ diffusivity, while the increase in PEO content increased CO₂ solubility. Operating at a gas feed pressure of 25 bar, 40 wt% polymer network-bearing membranes presented a CO₂ permeability of 272.8 Barrer and CO₂/N₂ selectivity of 79.6.

Silver ions in conjunction with UiO-66-NH₂ nanoparticles formed a precursor, which was used for the preparation of advanced mixed matrix membranes [87]. Under the operational conditions of CO_2/N_2 (10/90), under 25 °C and 2 bar, the membrane with 30%Ag+@10%UiO@PIM-1 composition achieved a 65% increase in CO_2/N_2 selectivity (near 30) and 120% increase in CO_2 permeability (>15,000 Barrer) compared to the pristine PIM-1 (polymer of intrinsic microporosity) membrane. With the optimization of the Ag⁺-doping concentration and Ag+@UiO-loading content, the CO_2/N_2 separation efficiency is greater than the 2019 upper bound. The combination of CO_2 facilitated transport, enhanced by the CO_2 -Ag⁺ affinity, and molecular sieving from pore regulation, due to the filling by Ag⁺ ions, has a synergic effect to achieve this CO_2/N_2 separation.

The introduction of ZIF-8 nanoparticles into Pebax-2533 is used to fabricate mixed matrix membranes [88]. The reaction of Zn and imidazole is the basis for the formation of ZIF-8 nanoparticles in the Pebax solution. Within a mixed membrane containing 8 wt% of nanofiller, there are increases of 28% (CO₂ permeability), 250% (CO₂/N₂ separation), and 208% (CO₂/CH₄ selectivity) with respect to the values reached by the use of the neat Pebax membrane. CO₂ permeated due the pore size and the affinity between CO₂ and ZIF-8 nanoparticles. This affinity was due to the electrostatic interactions with uncoordinated N of the mIm linker and unsaturated Zn sites of the nanoparticles.

A cobalt-based zeolite imidazole framework (ZIF-67) merged into poly(ether-blockamide) (Pebax-2533) resulted in a series of mixed matrix membranes [89]. The results showed that the CO₂ permeation increased from 63.8 Barrer (neat membrane) to 190.5 Barrer (16 wt% of ZIF-67). Operating at 25 °C and 4 bar, CO_2/N_2 and CO_2/CH_4 selectivities increased from 25.3 to 11.4 (neat membrane) to 39.7 and 22.5 (mixed membrane), respectively.

 CO_2 capture by membranes was modeled using the superstructure method [90]. With 96% CO_2 purity, the gas recovery efficiency was 90%, however, the combination of different membrane types increased the results. Over all the situations, a two-stage membrane process with recycling produced the best results by the use of the combination of a CO_2 -selective membrane and H₂-selective membrane. This operational form also had economic advantages compared with the Selexol process. It is claimed that under the best conditions, the CO_2 capture cost can be reduced to USD 11.75/t CO_2 .

A mixed matrix membrane based on poly(ether-block-amide) was formed with synthesized ZIF-8 nanoparticles embedded into a maltitol-modified Pebax[®]-1657 polymer [91]. The addition (0–10 wt%) of ZIF-8 nanoparticles into the Pebax/maltitol (20 wt%) matrix formed the various Pebax/maltitol/ZIF-8 mixed matrix membranes. At 30 °C and 10 bar, Pebax/maltitol (20 wt%)/ZIF-8 (10 wt%) membrane showed the highest (429.57 Barrer) CO₂ permeability, whereas the highest selectivity values of 69.31 CO₂/N₂ and 26.59 CO₂/CH₄ were obtained by Pebax/maltitol (20 wt%)/ZIF-8 (5 wt%).

CFA-based ceramic membrane was fabricated by depositing SiO₂ nanoparticles and grafting 1H, 1H, 2H, 2H-perfluorooctyltriethoxysilane (POTS) [92]. This modified ceramic membrane was used to capture CO_2 using ethanolamine as the absorbent. At 0.2 wt% drafting with POTS, the membrane had a maximum capture efficiency of almost 98% (Table 8) and a mass transfer rate of 21.17 mol/m²·h.

Membrane	0.2	0.4	0.6	0.8	1.0
Original	62	55	45	35	30
Superhydrophobic	98	90	75	67	62

Table 8. Influence of the gas flow rate (L/min) on CO₂ capture efficiency (%).

Absorbent flow rate: 0.1 L/min. Data from [92].

A nanofiber membrane containing iron oxide nanoparticles (NPsFe₂O₃) was prepared, and this was coupled to microalgae (*Chlorella vulgaris*) to improve CO₂ removal [93]. When the nanofiber membrane contained 4% NPsFe₂O₃, CO₂ dissolution increased. When the nanofiber membrane was used as a CO₂ adsorbent with semifixed microalgae culture (two layers), the results showed that with the culture, the biomass productivity, CO₂ fixation efficiency, and carbon fixation efficiency increased by 1.4 times with respect to the use of the pristine membrane (zero layers). CO₂ fixation was attributable to diverse factors: (i) physical adsorption by the pores, (ii) electrostatic gravity, (iii) van der Waals force combination, and (iv) Lewis acid–base effect.

An investigation used a hollow fiber membrane contactor to demonstrate the difference between water and air as sweeping media for CO₂ exchange [94]. When the aqueous CO₂ concentration is higher than 3.53×10^{-6} mL CO₂/L, air removes the gas better than water, however, at a lower gas concentration, the use of water is beneficial with respect to air.

The hydroxylation of MXene produced alkaline MXene nanosheets (TCOH), and this product together with Pebax-1657 polymer fabricated a mixed matrix membrane [95], while ionic liquid (IL) 1-ethyl-3-methylimidazolyl acetate ([EMIm][AcO]) was nanoconfined in the 2D channel of an ultrathin TCOH@Pebax-1657 membrane to make a composite membrane. The TCOH@Pebax-1657/IL membrane has convenient CO₂ permeation and selectivity values of 59.1 (CO₂/N₂) and 52.9 (CO₂/CH₄) at the ionic liquid concentration of 35 wt%.

Pyrolysis of Fe₃O₄@ZIF-8/polyimides resulted in the formation of hybrid carbon sieve membranes [96], with a permeability of 5130 Barrer and CO_2/N_2 and CO_2/CH_4 selectivities of 29 and 48, respectively. The separation of CO_2 from methane and nitrogen relies on molecular sieving due to the membrane structure, coupled with convenient CO_2 surface diffusion. Other features of these membranes are fast CO_2 diffusion rate and 30 days of resistance under operational conditions.

The joint use of 4,4'-oxydiphathalic anhydride and Jeffamine monomers produced different poly(amic acids) of different molecular weights [97]. These were used to fabricate silver nanoparticles via the chelating reaction with silver ions. The objectives of this methodology are (i) fabrication of mixed matrix membranes without defects, (ii) more efficient CO_2 selective permeation, and (iii) increases in the membrane's long-term usefulness. The inclusion of 2 wt% silver nanoparticles in the Pebax-1657 polymer increases CO_2 permeability by 50% and CO_2 selectivity by 100% with respect to the values derived from the use of the neat polymer. The presence of silver nanoparticles in the membrane was the key to improve the CO_2 diffusivity and the CO_2/CH_4 diffusivity–separation; also,

this physical process was accompanied with the facilitated transport of the CO_2 molecules by means of the nanoparticles.

Membrane contactors in catalyst-aided solvent regeneration were employed to remove CO_2 from industrial flue gas [98]. A ceramic membrane module was built to investigate this approach using as the solvent aqueous monoethanolamine (MEA) and powdered metatitanic acid [TiO(OH)₂] as the catalyst. The results showed that the nanofluid characteristics of the catalyst decreased the mass transfer resistance and enhanced the concentration change in the liquid film, which resulted in a greater CO_2 absorption rate. The CO_2 removal process was due to the reaction of CO_2 with MEA+catalyst, followed by the catalytic desorption at 90 °C, releasing the gas and leaving the MEA+catalyst mixture free of the gas. A simulation modeling an industrial-scale process resulted in a capture cost of USD 46.7/t CO_2 , a value higher than that given in [90], though they were obtained under different conditions.

Transport channels in graphene oxide membranes were incorporated by swift heavy ion irradiation [99]. After ¹²⁹Xe irradiation, typical ion tracks appeared within the graphene oxide membrane cross-section. The irradiated membranes increased the upper-limit H_2/CO_2 performance, with an increase in H_2 permeation of nearly three orders of magnitude.

A series of F-Ln (Ln = La, Ce, Pr, Nd) nanosheets with mesoporous structure were fabricated as a filler in Pebax[®]-1657 matrix [100]. The pore diameters of the nanosheets were in the following order: F-La > F-Ce > F-Pr > F-Nd, and a smaller pore diameter of nanosheets improved the CO₂ separation efficiency. Under dry conditions (Figure 6), the Nd membrane presented the highest CO_2/CH_4 selectivity value, whereas this tendency is also maintained under humidified conditions, in which Pebax/F-Nd-6 membrane showed CO_2 permeation up to 1265 Barrer and CO_2/CH_4 selectivity of 36.7, which represented increases of 2.3 times and 1.9 times with respect to the values presented by pure Pebax. After 120 h of operation, the separation of CO_2/CH_4 using the membranes remained near constant.



Figure 6. CO_2/CH_4 selectivities at F-Ln (Ln: 6 wt%). Feed pressure: 2 bar. Temperature: 25 °C. CO_2/CH_4 volume ratio (dry conditions): 30/70. Data from [99].

Polyethyleneimine (PEI) (PEI-F-Ce) and PEI-F-Ce/polyethylene oxide (PEO) mixed matrix membranes were fabricated to investigate their performance in CO₂ permeation [101]. An electrostatic self-assembly procedure was used to prepare PEI-F-Ce nanosheets at various PEI concentrations. The membranes loaded with 2% PEI-F-Ce-2.5 nanosheets presented CO₂ permeation of 641 Barrer and CO₂/N₂ selectivity of near 70. These values were better than those presented by pure PEO membranes by 62% and 53%, respectively, exceeding the 2019 upper bound. PEI-F-Ce-2.5 nanosheets, with a stable

pore channel of 3.406 nm, showed a CO_2 adsorption loading of 1.32 mmol/g, improving CO_2/N_2 selectivity. Furthermore, the reversible reaction between CO_2 and $-NH_2$ groups increased, at -20 °C, the CO_2/N_2 selectivity 5.1 times, broadening their use in cryogenic gas separation developments. The high porosity shown by PEI-F-Ce nanosheets was the key factor for CO_2 removal, whereas the in situ functionalization assisted in the interfacial compatibility with the membrane matrix. This also improved the CO_2/N_2 selectivity of the membrane.

Table 9 summarizes the nanomaterials and objectives of the above references.

Table 9. Summary of the applications of nanomaterials in the use of membranes to investigate CO₂ permeation and selectivity.

Ref.	Nanomaterial	Objective
[79]	PMP+metal oxides	Modeling CO ₂ permeation
[80]	Zeolite+Pebax-1657	CO_2/N_2 , CO_2/O_2 , CO_2/CH_4 separation
[81]	PEBA	CO_2 capture
[82]	KAUST-8 nanosheets	CO_2 uptake, CO_2/N_2 separation
[83]	Nanofillers+PEI	H_2/CO_2 separation
[84]	Zr-MOFs+AA+CS	CO_2 permeation, CO_2/N_2 separation
[85]	Graphene oxide+melanine	As above
[86]	PEO+SiO ₂ +Pebax1657	CO_2 diffusion, CO_2/N_2 separation
[87]	Silver+UiO66	CO_2/N_2 separation
[88]	ZIF-8+Pebax-2533	CO_2/N_2 , CO_2/CH_4 selectivity
[89]	ZIF-67+Pebax-2533	As above
[90]	Superstructure method	Modeling CO ₂ capture
[91]	ZIF-8+Pebax-1657	CO_2/N_2 , CO_2/CH_4 separation
[92]	Ceramic membranes	CO ₂ capture
[93]	Fe ₂ O ₃ +microalgae	CO_2 fixation efficiency
[94]	HFM contactor	CO_2 exchange in air or water
[95]	TCOH+Pebax-1657+IL	CO_2/N_2 , CO_2/CH_4 separations
[96]	Carbon membranes	As above
[97]	Silver+Pebax-1657	CO_2 permeation
[98]	Membrane contactor	CO ₂ removal from industrial flue gas
[99]	Graphene oxide	CO ₂ removal
[100]	Ln+Pebax-1657	CO_2/CH_4 selectivity
[101]	PEI+cerium	CO_2/N_2 separation

PMP: poly(4-methyl-1-pentene. PEBA: poly(ether-block-amide). PEI: polyetherimide. MOFs: metal–organic frameworks. AA: activated carbon. CS: chitosan. PEO: polyethylene oxide. HFM: hollow fiber module. TCOH: alkaline MXene nanosheets. IL: ionic liquid. Ln: lanthanides.

The above summary indicates that many of the references were dedicated to investigating the separation of CO_2 from N_2 and CH_4 , while just two used computational modeling to investigate CO_2 permeation and just one (ref. [98]) utilized in its experimentation a real industrial flue gas. Taking into account the same considerations as in Table 5, Table 10 presents absolute CO_2/N_2 and CO_2/CH_4 selectivity values derived from the use of nanomembranes.

Table 10. Selectivity values for CO_2/N_2 and CO_2/CH_4 separations using nanomembranes.

Ref.	CO ₂ /N ₂	Ref.	CO ₂ /CH ₄
 [86]	79.6	[92]	52.9
[84]	71.3	[96]	48.0
[101]	70.0	[100]	36.7
[87]	30	[91]	26.6
[84]	29.4	[89]	22.5
[96]	29		

Separation values may be dependent on pressure, temperature, membrane pore size, and gas (CO₂, N₂, and CH₄) composition.

As was previously mentioned with the results derived from the use of the other two technologies, there was a great disparity in the results derived from the use of nanomembranes for CO_2 capture.

5. Conclusions

This review summarized the latest (as of 2023) advances in technologies used for CO_2 separation applications. From the above, just one reference (ref. [98]) utilized a real industrial gas sample in the experimentation, and just three used computational modeling to investigate CO_2 capture. This means that the great majority of the works were of an experimental nature. Also, it is worth noting here that there are a reasonable number of investigated the usefulness of their respective nanomaterials in CO_2 capture in continuous cycles. Due to the disparity in the results, i.e., Tables 5 and 9, it is risky to ascertain, at first glance, which nanomaterial is the best in their respective technologies.

The practical experience of CO_2 capture by absorption methodologies makes it the most mature available technology. However, there is still space for further improvements in order to reduce costs, including capital and operation. Investigations on avoiding solvent degradation and enhancement of the solvent's regenerative efficiency are needed in order to develop small-scale, commercially viable direct air capture plants. Also, it is of the utmost necessity to develop newer absorbents, increasing the overall CO_2 removal efficiency. Of current (and future) interest is the investigation of the use of porous liquids in CO_2 capture, with studies on the construction of stable and structurally diverse porous liquids that can achieve efficient developments in this research field.

Efforts in adsorption processing must be directed at the enhancement of the properties of the adsorbent material in order to improve CO_2 uptake on them; these enhancements may be investigated by developments of thermal, acid, transition metal, and organic modifications. Nanomaterials, with their clear advantages, i.e., elevated CO_2 adsorption efficiency, remarkable CO_2 separation from other gases, adequate adsorption kinetics, and proper recyclability under different environments, have a series of weak points in their uses: the relative lack of chemical stability of metal–organic frameworks in aqueous solution, elevated POP costs, and mesoporous materials presenting a low adsorption loading at low pressure. Obviously, these will be the focus of future investigations to improve these weaknesses.

Membrane-based CO_2 separation needs developments to improve the technology with respect to stability, scalability, separation properties, and costs against effectiveness of the different membranes used in the operations. Also, the weakness in membrane permeability and mechanical strength will be other point to improve. The incorporation of fillers into the polymer matrix, to form mixed matrix membranes, must to comply with these improvements (permeability, selectivity, and plasticization) by modulation of their chemical properties, thus widening the use of these membranes to a greater scale. The above, together with the use of better absorbents, will increase the use of these membranes in CO_2 capture.

Another point of future interest will be the implantation of these advanced nanomaterial technologies for CO_2 recovery in cryogenic circuits to improve the efficiency of the whole process.

Also, the use of nanomaterials in real gas streams needs to be investigated since most of the references (all but ref. [98]) used mimic gas streams. The real performance of the above technologies coupled with the use of nanomaterials has to be proven in real situations, which, as one can realize, may be quite different to that found in laboratory-directed investigations and those using mimic gases.

Table 11 summarizes, in the opinion of the present author, pros and cons of the technologies reviewed in the manuscript and used in CO_2 capture.

Technology	Pros	Cons
Absorption	Established technology	Chemistry of amines, regeneration of solvent, stability of the adsorbent
Adsorption	Established technology, elevated gas removal capacity	Possible generation of toxic wastes, stability of the adsorbent
Membranes	Modular configuration, adequate surface area per unit volume	Limitations due to gas permeation, resistance due to degradation of membrane

Table 11. Pros and cons of the technologies used in CO₂ capture.

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